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13. ABSTRACT (Maximum 200 words) The research in this proposal focused on the development of femtosecond laser techniques and their applications in the studies of molecular dynamics in real time. The research culminated in some forty publications with the involvement of more than twenty-five graduate students, post-doctoral fellows, and visiting associates from the U.S. and abroad.			
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## FINAL TECHNICAL REPORT

AFOSR Grant No. F49620-97-1-0227  
ULTRAFAST DYNAMICS OF CHEMICAL REACTIONS

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### SUMMARY OF WORK ACCOMPLISHED

In this section, we highlight the research accomplishments of the work supported by this grant by summarizing the findings presented in some forty publications of the Caltech group, with the references given in every section.

#### (A) PROBING METHODS

In the previous grant, we outlined the method used for probing the transient behavior of dissociation reactions and reactions restricted to van der Waals complexes (so-called "oriented" bimolecular). In most cases, the probing method depends on the change in the laser-induced fluorescence as fragments separate. Since the recoil velocity of fragments is typically 1 km/second, the current state of the art in femtosecond technology (6 fs duration) allows one to view motions, in principle, with a resolution of  $\sim 0.06 \text{ \AA}$ .

We have made progress in developing the probing methodology in three new directions. First, by building a molecular beam system that can

be housed on a floating optical table with a time-of-flight mass spectrometer we were able to record changes in mass or mass distributions while maintaining the delicate femtosecond (fs) resolution. Moreover, we could resolve the velocity and angular distributions of fragments and study their fs time evolution. This so-called kinetic-energy-time-of-flight (KETOF) method has proven very useful in resolving the dynamics of complex systems, as described in the references given below.

Second, we have extended the range of probing to high energies and to higher spatial resolution in order to resolve trajectories of the dynamics in  $r$  and  $t$ , allowing one to plot the evolution of wave packets in a 2-D picture representing distance ( $r$ ) and time ( $t$ ) coordinates. This study was detailed in the full account given below.

Third, for systems probed under extreme environment, we found that nonlinear optical techniques have the required sensitivity. We employed degenerate four wave mixing, but now with fs resolution in the gas phase, to study unimolecular dissociation and bimolecular reactions at very low densities and also at higher pressures. In a comprehensive report published recently, we have given the theoretical description and operational approach for these different probing methods.

The new findings of the research done in these areas are discussed in the following publications:

1. Kinetic-Energy, Femtosecond Resolved Reaction Dynamics: Modes of Dissociation (in Iodobenzene) from Time-Velocity Correlations  
P. Y. Cheng, D. Zhong, and A. H. Zewail  
Chem. Phys. Lett., 237, 399 (1995)
2. Transition States of Charge-Transfer Reactions: Femtosecond Dynamics and the Concept of Harpooning in the Bimolecular Reaction of Benzene with Iodine  
P. Y. Cheng, D. Zhong, and A. H. Zewail  
J. Chem. Phys., 103, 5153 (1995)
3. Microscopic Solvation and Femtochemistry of Charge-Transfer Reactions: The Problem of Benzene(s)-Iodine Binary Complexes and Their Solvent Structures  
P. Y. Cheng, D. Zhong, and A. H. Zewail  
Chem. Phys. Lett., 242, 368 (1995)
4. Femtosecond, Velocity-Gating of Complex Structures in Solvent Cages  
P. Y. Cheng, D. Zhong, and A. H. Zewail  
J. Phys. Chem., 99, 15733 (1995)
5. Femtosecond Real-Time Probing of Reactions. XVIII: Experimental and Theoretical Mapping of Trajectories and Potentials in the NaI Dissociation Reaction  
P. Cong, G. Roberts, J. L. Herek, A. Mohktari, and A. H. Zewail  
J. Phys. Chem., 100, 7832 (1996)
6. Femtosecond Real-Time Probing of Reactions: XIX Nonlinear (DFWM) Techniques for Probing Transition States of Uni- and Bimolecular Reactions  
M. Motzkus, S. Pedersen, and A. H. Zewail  
J. Phys. Chem., 100, 5620 (1996)

7. Femtosecond Real-Time Probing of Reactions: XXII Kinetic Description of Probe Absorption, Fluorescence, Depletion and Mass Spectrometry  
S. Pedersen and A. H. Zewail  
Molecular Physics, Vol. 89, No. 5 1455 (1996)
8. Femtosecond real-time probing of reactions. 23. Studies of temporal, velocity, angular and state dynamics from transition states to final products by fs-resolved mass spectrometry  
D. Zhong and A. H. Zewail  
J. Phys. Chem., Submitted for publication

#### (B) ELEMENTARY AND COMPLEX REACTIONS

With the different fs techniques available in our laboratories, we were able to study elementary and complex reactions. These include studies of the scalar and vectorial (alignment) properties of reactions, the effect of intense laser fields on reaction dynamics and the role of centrifugal forces on the elementary dynamics of dissociation. For complex molecular systems, we were successful in utilizing the mass spectrometric approach (Section A) to study the dynamics of many organic and inorganic reactions: pericyclic, Norrish, isomerization, tautomerization, and charge transfer reactions, and others. The new findings are given in the following publications:

1. Femtosecond Real-Time Probing of Reactions: XV. Time-Dependent Coherent Alignment.  
J. S. Baskin and A. H. Zewail  
J. Phys. Chem., 98, 3337 (1994)

2. Femtosecond Real-Time Probing of Reactions: XVI. Dissociation with Intense Pulses  
A. Materny, J. L. Herek, P. Cong, and A. H. Zewail  
J. Phys. Chem., 98, 3352 (1994)
3. Femtosecond Real-Time Probing of Reactions: XVII Centrifugal Effects in Direct Dissociation Reactions  
G. Roberts and A. H. Zewail  
J. Phys. Chem., 99, 2520 (1995)
4. The Validity of The "Diradical" Hypothesis: Direct Femtosecond Studies of the Transition-State Structures  
S. Pedersen, J. L. Herek, and A. H. Zewail  
Science, 266, 1359 (1994)
5. Femtochemistry of Organometallics: Dynamics of Metal-Metal and Metal-Ligand Bond Cleavage in  $M_2(CO)_{10}$   
S. K. Kim, S. Pedersen, and A. H. Zewail  
Chem. Phys. Lett., 233, 500 (1995)
6. Direct Femtosecond Observation of the Transient Intermediate in the  $\alpha$ -Cleavage Reaction of  $(CH_3)_2CO$  to  $2CH_3 + CO$ : Resolving the Issue of Concertedness  
Sang Kyu Kim, Soren Pedersen, and A. H. Zewail  
J. Chem. Phys., 103, 477 (1995)
7. Femtosecond Elementary Dynamics of Transition States and Asymmetric  $\alpha$ -Cleavage in Norrish Reactions  
S. K. Kim and A. H. Zewail  
Chem. Phys. Lett., 250, 279 (1996)
8. Femtosecond Molecular Dynamics of Tautomerization in Model Base Pairs  
A. Douhal, S. K. Kim, and A. H. Zewail  
Nature, 378, 260 (1995)

9. Femtosecond Real-Time Probing of Reactions: XX. Dynamics of Twisting, Alignment, and IVR in the *trans*-Stilbene Isomerization Reaction  
J. S. Baskin, L. Bañares, S. Pedersen, and A. H. Zewail  
J. Phys. Chem., 100, 11920 (1996)
10. Femtosecond Real-Time Probing of Reactions: XXI Direct Observation of Transition-State Dynamics and Structure in Charge-Transfer Reactions  
P. Y. Cheng, D. Zhong, and A. H. Zewail  
J. Chem. Phys., 105, 6216 (1996)
11. Retro-Diels-Alder Femtosecond Reaction Dynamics  
B. A. Horn, J. L. Herek, and A. H. Zewail  
J. Am. Chem. Soc., 118, 8755 (1996)
12. Femtosecond Nucleophilic-Substitution ( $S_N$ ) Reaction Dynamics  
D. Zhong, S. Ahmad, P. Y. Cheng, and A. H. Zewail  
J. Am. Chem. Soc. 119, 2305 (1997)
13. Femtosecond Elimination Reaction Dynamics  
D. Zhong, S. Ahmad, and A. H. Zewail  
J. Am. Chem. Soc. 119, 5978 (1997)

### (C) BIMOLECULAR REACTIONS

We built on earlier studies of using van der Waals complexes of the two reagents to study the bimolecular dynamics of the reaction. We examined the system of halogen + alkyl halide reactions by starting from the precursor  $\text{CH}_3\text{I} \cdots \text{ICH}_3$ . With a fs initiation pulse the  $\text{CH}_3$  group leaves in  $\sim 100$  fs and we could study the inelastic I atom scattering and the reactive  $\text{I}_2$  production. The approach and the different reactions studied so far is described in the publications given below. We have also examined a

different type of bimolecular reaction. A CN radical was generated (from ICN) on the fs time scale and its reactions with bulk  $\text{CHCl}_3$  and  $\text{CH}_3\text{Cl}$  (solution) was studied following the transient absorption change with time. This way the processes of abstraction (to form HCN) and caging were studied in real time. The new findings are given in the following publications:

1. Bimolecular Reactions Observed by Femtosecond Detachment to Aligned Transition States: Inelastic and Reactive Dynamics  
D. Zhong, P. Cheng, and A. H. Zewail  
J. Chem. Phys., 105, 7864 (1996)
2. Dynamics of Ground State Bimolecular Reactions  
C. Wittig and A. H. Zewail  
(Oxford University, New York, 1996) p. 64 - Book Chapter
3. Femtochemistry of ICN in Liquids: Dynamics of Dissociation, Recombination and Abstraction  
Chaozhi Wan, Manish Gupta, and A. H. Zewail  
Chem. Phys. Lett., 256, 279 (1996)
4. Femtosecond Dynamics of a Hydrogen-bonded Model Base Pair in the Condensed Phase: Double Proton Transfer in 7-Azaindole  
M. Chachisvilis, T. Fiebig, A. Douhal, and A. H. Zewail  
J. Phys. Chem. 102, 669 (1998)

**(D) CONTROL AND STRUCTURAL IMAGING BY  
DIFFRACTION AND COHERENCE SPECTROSCOPY**

In the area of reaction control, we have focused our effort on two directions: the use of timed pulse sequences to change the branching ratio or yield of a reaction, and the use of fs chemical activation to "control" the



extent of IVR in complex molecular systems. In the former direction, we studied simple systems (such as NaI) and invoked different pulses to alter the yield in the two channels involved:  $\text{Na} + \text{I}$  and  $\text{Na}^* + \text{I}$ . In the case of chemical activation, we investigated Norrish-type reactions and showed a controlled nonstatistical behavior if activation is initiated with fs pulses (see below).

We have devoted a major effort to study structural changes of reactions using coherence spectroscopy and diffraction imaging. Rotational coherence spectroscopy, a time resolved method, is now well developed and has been successful in deducing structures of more than 120 complex molecular systems. Details can be found in the book chapter given below. For imaging, we have chosen the method of ultrafast electron diffraction (UED). In recent two publications we have provided our experimental and theoretical effort in this area using the Caltech first generation apparatus. We were able to generate ps electron pulses and to obtain gas phase diffraction patterns with this time resolution. In the coming three years, the focus is on the clocking of structural changes using the second generation apparatus which is part of our discussion in the coming section. The new findings are given in the following publications:

1. Femtosecond Control of an Elementary Unimolecular Reaction From the Transition-State Region  
J. L. Herek, A. Materny, and A. H. Zewail  
Chem. Phys. Lett., 228, 15 (1994)
2. Femtosecond Chemically-Activated Reactions: Concept of Non-statistical Activation at High Thermal Energies  
S. K. Kim, J. Guo, J. S. Baskin, and A. H. Zewail  
J. Phys. Chem., 100, 9202 (1996)
3. Molecular Structures from Ultrafast Coherence Spectroscopy.  
P. M. Felker and A. H. Zewail  
in: Femtosecond Chemistry, eds. J. Manz and L. Wöste (VCH Publishers, Inc., New York, 1994) - Book Chapter
4. Ultrafast Electron Diffraction. IV. Molecular Structures and Coherent Dynamics  
J. C. Williamson and A. H. Zewail  
J. Phys. Chem., 98, 2766 (1994)
5. Ultrafast Electron Diffraction. V. Experimental Time Resolution and Applications.  
M. Dantus, S. B. Kim, J. C. Williamson, and A. H. Zewail  
J. Phys. Chem., 98, 2782 (1994)
6. Clocking transient chemical changes by ultrafast electron diffraction  
J. C. Williamson, J. Cao, H. Ihee, H. Frey, and A. H. Zewail  
Nature 386, 159 (1997)
7. Ultrafast Electron Diffraction: Structures in Dissociation Dynamics of Fe(CO)<sub>5</sub>  
H. Ihee, J. Cao, and A. H. Zewail  
Chem. Phys. Lett. 281, 10 (1997)

## (E) GENERAL REPORTS

In several review articles from this group, or in collaboration with other colleagues, the above results and their general implications have been discussed (previous reviews related to this work were given in the last renewal grant):

1. Femtochemistry: Concepts and Applications  
A. H. Zewail  
in: Femtosecond Chemistry, eds. J. Manz and L. Wöste (VCH Publishers, Inc., New York, 1994) - Book Chapter
2. Ultrafast Dynamics of the Chemical Bond - Femtochemistry  
A. H. Zewail  
in: Ultrafast Processes in Chemistry and Photobiology, Chemistry for the 21st Century, eds. M. A. El-Sayed, I. Tanaka, and Y. N. Molin (IUPAC, Blackwell Scientific Publishers, Oxford, United Kingdom, 1994) - Book Chapter
3. Rotational Coherence Phenomena  
P. M. Felker and A. H. Zewail  
in: Jet Spectroscopy and Molecular Dynamics, eds. M. Hollas and D. Phillips (Chapman and Hall, Blackie Academic, 1994) - Book Chapter
4. Ultrafast Dynamics of IVR in Molecules and Reactions  
P. M. Felker and A. H. Zewail  
in: Jet Spectroscopy and Molecular Dynamics, eds. M. Hollas and D. Phillips (Chapman and Hall, Blackie Academic, 1995) P. 222 - Book Chapter
5. Transient Species at Femtosecond Resolution  
A. H. Zewail  
Proceedings of The Robert A. Welch Foundation 38th Conference on Chemical Research, Chemical Dynamics of Transient Species, Houston, Texas (1994), p. 129

6. Coherence -- A Powerful Concept in the Studies of Structures and Dynamics  
A. H. Zewail  
Laser Physics, 5, 417 (1995)
7. Direct Observation of The Transition State  
J. C. Polanyi and A. H. Zewail  
Accounts of Chemical Research (Holy-Grail Special Issue), 28, 119 (1995)
8. Femtosecond Dynamics of Reactions: Elementary Processes of Controlled Solvation  
A. H. Zewail  
Berichte der Bunsengesellschaft für Phys. Chem., 99, 474 (1995)
9. Femtochemistry and Max Bodenstein's Impact  
A. H. Zewail  
Gas Phase Chemical Reaction Systems, 100 Years After Max Bodenstein, J. Wolfrum *et al.* (Eds.), Springer Series in Chemical Physics, New York, Volume 61, P. 3 (1996)
10. Femtochemistry -- Advances Over a Decade  
A. H. Zewail  
Femtochemistry -- Ultrafast Chemical and Physical Processes in Molecular Systems, M. Chergui (Ed.), World Scientific, Singapore (1996), p. 3
11. Proton-transfer Reaction Dynamics  
A. Douhal, F. Lahmani, and A. H. Zewail  
Chem. Phys., Special Issue, 207, 477 (1996)
12. Femtochemistry: Recent Progress in Studies of Dynamics and Control of Reactions and Their Transition States  
A. H. Zewail  
J. Phys. Chem., Centennial Issue, 100, 12701 (1996)
13. Femtochemistry: Dynamics with Atomic Resolution  
A. H. Zewail  
in Femtochemistry & Femtobiology, V. Sundström (Ed.), World Scientific, Singapore (1997)

14. Femtochemistry--Atomic-Scale Resolution of Physical, Chemical and Biological Dynamics

A. H. Zewail

The Robert A. Welch Foundation (Chemistry Award Paper)  
Proceedings of The 41st Conference on Chemical Research,  
The Transactinide Elements, Houston, Texas (1997), p. 32<sup>3</sup>

**THESES COMPLETED BY ZEWEIL RESEARCH GROUP**  
**IN LAST TWO YEARS**

(Copies Available in the California Institute of Technology  
Archives and the Chemistry Division Library)

1.    **Soren Pedersen**  
      **Development and Applications of Techniques in Laser**  
      **Femtochemistry**  
      Submitted February 5, 1996 - Awarded February, 1996  
      Present address:       McKinsey & Company, Inc.  
                              Ved Stranden 14  
                              DK-1061 Copenhagen K, Denmark
  
2.    **Jennifer L. Herek**  
      **Femtochemistry and Reactive Intermediates; Application to**  
      **Atmospheric and Organic Chemistry**  
      Submitted June 6, 1996 - Awarded June, 1996  
      Present address:       Chemical Physics  
                              Lund University  
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                              S-221 00 Lund, Sweden
  
3.    **Qianli Liu**  
      **Femtosecond Real-Time Dynamics of Solvation: Molecular**  
      **Reactions in Clusters and Supercritical Fluids**  
      Submitted August 12, 1996 - Awarded August, 1996  
      Present address:       1160 East Cordova Street, Apt. 3,  
                              Pasadena, California 91106  
                              (Affiliated with Pasadena Computers)
  
4.    **Chuck Williamson**  
      **Ultrafast Gas-Phase Electron Diffraction**  
      Submitted August 21, 1997 - To be awarded June 1998  
      Present address:       632 Westgate Street, Apt. 38  
                              Iowa City, Iowa 52246  
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# BIOGRAPHICAL SKETCH

Ahmed H. Zewail

Ahmed Zewail currently is the Linus Pauling Professor of Chemistry and Professor of Physics at the California Institute of Technology, and Director of the NSF Laboratory for Molecular Sciences (LMS). He received his B.S. and M.S. degrees from Alexandria University, and his Ph.D. from the University of Pennsylvania. After the completion of his Ph.D., he went to the University of California, Berkeley, as an IBM research fellow. Zewail was appointed to the faculty at Caltech in 1976. In two years he was tenured, in 1982 became a full professor, and in 1990 was honored by the first Linus Pauling Chair at Caltech.

Zewail holds *honorary degrees* from the American University, Cairo, Oxford University, Katholieke University (Leuven, Belgium), University of Pennsylvania (USA), and Université de Lausanne. He gave more than one hundred named and plenary lectures, and he has been the John van Geuns Stichting Professor at University of Amsterdam, Rolf Sammet Professor at Johann Wolfgang Goethe University and Christensen Professorial Fellow at Oxford. He served as Visiting Professor at the University of Bordeaux, Ecole Normale Supérieure, University of California, Los Angeles, American University, Cairo, Texas A&M, University of Iowa, and Collège de France.

Ahmed Zewail has been widely recognized for his contributions. His honors include the Robert A. Welch Prize Award, Wolf Prize, King Faisal Prize, Benjamin Franklin Medal, Leonardo Da Vinci Award of Excellence, Röntgen Prize, Paul Karrer Gold Medal, Bonner Chemiepreis, Medal of the Royal Netherlands Academy of Arts and Sciences, Carl Zeiss Award, Hoechst Award, and the Alexander von Humboldt Award. From the American Physical Society, he was awarded the Herbert P. Broida Prize and the Earle K. Plyler Prize, and from the American Chemical Society, the Nichols Medal, Linus Pauling Medal, E. Bright Wilson Award, Peter Debye Award, Nobel Laureate Signature Award, Harrison-Howe Award, and the Buck-Whitney Medal. From the National Academy of Sciences, the Chemical Sciences Award, and from Yale University, the J. G. Kirkwood Award. In 1995 he received the Order of Merit, first class. He was Alfred P. Sloan fellow, Camille and Henry Dreyfus Teacher-Scholar, and John Simon Guggenheim fellow.

Zewail is a member of the National Academy of Sciences, American Academy of Arts and Sciences, Third World Academy of Science, European Academy of Arts, Sciences and Humanities, and fellow of the American Physical Society.

Born February 26, 1946, he has a family of four children and is married to Dema Zewail, a physician in public health (UCLA). His scientific family over the past 20 years consists of some 120 graduate students, post-doctoral research fellows and visiting associates. He lives in San Marino, California.

Zewail's current research is devoted to ultrafast lasers and electrons and applications in chemistry and biology. In the field of femtochemistry, developed by the Caltech group, the focus is on the dynamics of elementary femtosecond ( $10^{-15}$  second) processes in the gas phase, in clusters and in the condensed phase.

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